948,870

PATENT SPECIFICATION

NO DRAWINGS

948,870

Date of Application and filing Complete Specification: Oct. 21, 1958.
No. 33575/58.
Application made in Germany (No. W22082 IVa/78c) on Oct. 22, 1957.
Complete Specification Published: Feb. 5, 1964.

Index at acceptance:—C1, D6.

International Classification:—C 06 b.

COMPLETE SPECIFICATION

Improvements in or relating to Propellants

We, WASAG-CHEMIE AKTIENGESELLSCHAFT, of 9, Rolandstrasse, Essen, Germany, a body corporate organised under the Laws of Germany, do hereby declare the invention for which we pray that a patent may be granted to us, and the method by which it is to be performed, to be particularly described in and by the following statement:—

This invention relates to propellants, principally for use in projectiles and rockets, which though burning at relatively low temperatures give a better performance than conventional propellant powders.

As known, conventional propellants, as used in artillery and small arms ammunition and for the propulsion of rockets, derive their energy from a reaction between oxygen, carbon and hydrogen. This applies to ordinary gunpowder as well as to the various modern types of powder propellants, such as perchlorate powders, single-base powders, double-base and triple-base powders, as the fundamental principle of their combustion is not affected by whether the oxygen is supplied by a substance which has been mechanically mixed with the powder or whether, in colloidally bound form, it is directly available in the powder as is the case in nitrocellulose powders, or whether it is actually present in the molecule itself of a chemically homogeneous propellant. The decisive point is the presence of the oxygen in a thermally unstable form from which it is readily released into closest possible proximity with suitable "acceptors".

In course of time propellants composed in this manner have been constantly further improved to meet the rising demands made upon them by the continuous development of better arms, the improvements having been due to additives or variations in the proportions of their components. Nevertheless, the results still fall short of requirements although the possibilities of further improvement in their performance appear to have been well-nigh exhausted. Another critical factor is the stability of such powders and of the constituents that enter into their composition. A large number of tests have been devised to ensure a satisfactory measure of safety when such powders are stored for long periods.

The term "performance" used in this Specification in relation to a propellant is intended to denote the conversion of energy of the propellant into velocity of the combustion products and is expressed in metres per second.

Past developments may here be briefly outlined. The desire to improve the performance of a propellant without increasing the size of the charge necessarily involved the development of powders with an increasing content of oxygen. This rise in oxygen content caused a concomitant increase in the temperatures developed in the explosion because larger amounts of heat were liberated in the exothermic reaction. The life of gun barrels depends upon the temperature of the gases of decomposition so that this development in the direction of ever "hotter" powders which had been pursued in various ways for a considerable period of time eventually proved to fall short of expectations. Owing to the incessant call for higher and higher performance and the consequent increase in the wear and tear of the barrels, recourse was had to the production of "cold" powders which at relatively low temperatures (inter alia by raising the volume of the gases that were evolved) also released considerable energies. Examples of this type of powder are the diglycol and nitroguanidine powders. However, both theory and practice indicate that the performance that can be achieved by this method cannot exceed a ceiling of about 1100 m/sec.

The present invention contemplates approaching the problem of providing propellants of higher performance along entirely different lines in which the disadvantages inherent in conventional powders will not arise.

According to the present invention there is provided a propellant comprising an oxygen-free endothermic compound, which, if it is a hydrocarbon has a C:H ratio of less than 1, and which yields decomposition products in the form of one or more gases having gas

constants as hereinafter defined), higher than 40, together with a conventional propellant and/ r explosive which contains more than one carbon atom in the molecule and which has a balanced oxygen content or an oxygen deficit, the endothermic compound constituting at least 20% by weight of the propellant.

The term "gas constant" as employed in this Specification is not to be confused with the molar gas constant R defined by the gas law $PV=RT$ but is in fact the molar gas constant divided by the molecular weight of the gas and is measured in kg. m. $\times$ degrees^{-1} $\times$ kg.$^{-1}$.

It is desirable that the gases evolved should have not only gas constants higher than 40, but should also contain low numbers of atoms so as to have correspondingly high values of kappa k i.e. the ratio of the specific heat at constant pressure c_p to that at constant volume c_v.

The present propellants preferably comprise at least 30% by weight of the oxygen-free endothermic compound, and may, if desired, comprise two or more of the endothermic compounds, possibly with the addition of a conventional stabilizer, such as urea or an aromatic substitution product thereof.

The components envisaged by the invention require an external activator to induce them to release their energy by decomposing. A high energy nitroglycerine powder has been found to provide a suitable activating charge, a normally primed small quantity being sufficient to provide the necessary energy of activation for the decomposition of the endothermic component. However, other powders may also be used, depending upon the varying decomposition temperatures of different components. Another possibility is to derive the energy necessary for activation from one of the usual primers, such as mercuric fulminate alone or mixed with potassium chlorate, or lead azide. By the selection of a suitable type of activation the velocity of decomposition is capable of being controlled.

Especially useful propellant components constituted in the manner proposed by the present invention are, in the first place, unsaturated aliphatic hydrocarbons. For instance, 1.3-butadiene offers advantages, as does cyclopentadiene, whereas acetylene lacks the required stability and must be excluded. Further, boron hydrides, lithium hydrides, lithium-boron hydrides, and other metal hydrides, as well as anhydrous hydrazine are useful propellant components which satisfy the invention. Especially their higher homologues are easy to handle because they are solids. An improvement in performance is also achieved if an unsaturated cyclic compound consisting principally of carbon and hydrogen, such as pinene, carbozile or diphenylamine is employed as a propellant component.

All these substances have in common that they will not decompose unless they are suitably activated and the propellants are therefore easily prepared. Neither their production nor their transportation or storage need be subjected to the same stringent rules which normally make the safe handling of explosives so inconvenient. Also in this respect the propellants proposed by the present invention therefore differ to their advantage from the conventional explosives hitherto used.

The table appended hereunder gives data relating to a "hot" and a "cold" powder compared with propellants constituted according to the invention.

The processing of propellants according to the invention depends upon whether the components are solids, liquids, or gases, when processed. Substances which at normal temperatures are gases may conveniently be processed at temperatures below their boiling points, for instance in the case of 1,3-butadiene at temperatures below $-5°$ C. at which the 1.3-butadiene is liquid.

The preparation of a propellant charge comprising a liquid component may be illustrated in the case of cyclopentadiene:—A basic charge of 30% by weight of "hot" nitroglycerine powder (N.G.P.) with an energy content of about 1250 Cal/kg is placed into the bottom of a cartridge case and covered with a sealing plate of N.G.P. over the threaded percussion primer. 70% by weight of liquid cyclopentadiene is then poured into the case, filling the space that remains above the N.G.P. Care must be taken to see that the projectile forms an airtight seal in the cartridge.

Propellants with solid energy-supplying components, such as carbazole, diphenyl-amine, and higher homologues of boron hydrides and B-Li-hydrides, are preferably shaped and profiled, this process affording a means of controlling the combustion velocity as may be desired. They are especially suitable as propellant charges for rockets.

Propellants prepared according to the invention may also be used for other purposes than those which have been discussed. For instance, their application as explosives in the manner of "Sprengel solutions" gives excellent results, as their energy content is very high.

As known, the work done by a propellant is actually performed by the highly compressed gases evolved in combustion. It is therefore an advantage if these gaseous combustion products are of a kind that will best meet the requirements of the intended purpose and which have properties that are best suited thereto. The performance of a propellant, i.e. its interior ballistic efficiency—disregarding other interior ballistic relationships—is a function of the flow velocity of the combustion gases inside the barrel, or inside the nozzle of

a rocket respectively, and therefore rises and falls in proportion with sonic velocity a in conformity with the equation

$$a = \sqrt{g \cdot \kappa \cdot RT}$$

(a = sonic velocity in $m.sec^{-1}$; g = acceleration of gravity in $m.sec^{-2}$, $\kappa = c_p/c_v$, R = gas constant in $kg.m\ degree^{-1} . kg^{-1}$, T = temperature in °C).

Conventional propellants generally decompose into CO, CO_2, H_2 and N_2, the "hot" powders especially evolving many tri-atomic gases with low kappa values. For a given oxygen content of the powder the ratio between the gaseous components CO_2, CO, H_2O, H_2 is determined by the law of mass action, that is to say it depends upon the water-gas equilibrium at the prevailing temperature. The better the oxygen balance of a powder the more CO_2 and H_2O will form. The gas constants R have the following values

$$CO = 30.28,\ CO_2 = 19.25,\ H_2O = 47.10,\ H_2 = 420.75.$$

The invention therefore aims at increasing especially the H_2 content of the combustion gases of the propellant above that of conventional propellants. Experimental data have established the favourable properties of H_2 as a gaseous propellant. During the second world war a so-called "hydrogen-gun" was constructed which gave a substantial increase in performance. However, owing to its considerable complexity, such a weapon is at present still an impracticable proposition.

Owing to its very low specific heat and its low atomic weight the carbon that is released has very favourable flow properties inside the barrel. However, the presence of other reaction products with a low atomic weight, such as Li or B in the combustion gases also substantially contributes to better performance. All this sheds a vivid light upon the inherent possibilities of the improvement in performance that may be obtained when using propellants composed as proposed by the present invention.

Powder Type	Heat Content at Kg.cal/Kg. (calculated constant volume)	Temp. °C.	Gas volume litre/kg (measured)	κ	R $\frac{\text{kg.m}}{\text{degree kg}}$	κ.R	κ.R.T
Hot powder	685	2010	998	1.26	37.82	47.6	95,300
Cold powder	542	1560	1112	1.28	41.3	52.8	82,500
40% N.G.P. 60% butadiene	678	1435	1060	1.27	58.0	73.8	105,500
20% N.G.P. 80% butadiene	814	1195	1150	1.37	92.7	127.1	151,800
40% N.G.P. 60% Cyclopentadiene	722	1340	992	1.27	53.9	68.8	91,300
40% N.G.P. 60% hydrazine	642	1395	1560	1.31	59.2	77.2	108,000
40% N.G.P. 60% carbazole	764	1685	710	1.245	44.6	55.7	93,800
50% N.G.P. 50% diphenylamine	708	1610	770	1.25	45.0	56.2	90,500

Note: The values of R for the several components are taken from the reference book "Hütte" (published by Wilhelm Ernst & Sohn, Berlin) the R-values for water and carbon are calculated, carbon being taken as gaseous. The specific heat of carbon at 800° C. is about 4, but to make ample allowance for the temperatures listed above the specific heat of carbon was taken as 6.

WHAT WE CLAIM IS:—

1). A propellant comprising an oxygen-free endothermic compound, which, if it is a hydrocarbon has a C:H ratio of less than 1, and which yields decomposition products in the form of one or more gases having gas constants, (as hereinbefore defined), higher than 40, together with a conventional propellant and/or explosive which contains more than one carbon atom in the molecule and which has a balanced oxygen content or an oxygen deficit, the endothermic compound constituting at least 20% by weight of the propellant.

2). A propellant as claimed in Claim 1, in which the oxygen-free endothermic compound is an unsaturated aliphatic hydrocarbon, anhydrous hydrazine, a boron hydride or other metal hydride or an unsaturated cyclic compound consisting principally of carbon and hydrogen.

3). A propellant as claimed in Claim 2, in which the unsaturated aliphatic hydrocarbon is 1,3-butadiene or cyclopentadiene.

4). A propellant as claimed in Claim 2, in which the unsaturated cyclic compound is pinene, carbazole or diphenylamine.

5). A propellant as claimed in any preceding Claim, in which the propellant comprises at least 30% by weight of the oxygen-free endothermic compound.

6). A propellant as claimed in any preceding Claim, in which the propellant comprises two or more of said endothermic compounds.

7). A propellant as claimed in any preceding Claim, in which the propellant has an addition of a conventional stabiliser.

8). A propellant as claimed in any preceding Claim, in which the propellant is activated by a firing charge consisting of nitroglycerine powder.

9). A propellant as claimed in any one of Claims 1 to 7, in which the energy of activation is supplied by a primer.

10). A propellant constituted and prepared substantially as hereinbefore described and exemplified.

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Leamington Spa: Printed for Her Majesty's Stationery Office, by the Courier Press (Leamington) Ltd.—1964. Published by The Patent Office, 25 Southampton Buildings, London, W.C.2, from which copies may be obtained.